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ATOMISTIC SIMULATION OF DEFECT PROPERTIES IN BCC TANTALUM

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The fundamental atomic-level properties of point and line defects in bcc Ta have been simulated by means of quantum-based multi-ion interatomic potentials derived from the model generalized pseudopotential theory (MGPT). The potentials have been applied to the calculations of point defect formation and migration energies. The results are then compared with the *ab-initio* electronic-structure results and experimental data, which in turn provide rigorous validation tests of the MGPT potentials. Robust and accurate two- and three-dimensional Green's function (GF) techniques have been developed for static and dynamic simulations of single a/2<111> screw dislocation properties in bcc Ta. The transformation of the dislocation core under the influence of external stress was studied in detail using static GF method. Finite-temperature GF simulation reveals multiple-kink (thermal-kink) formation under an applied stress and the corresponding thermal-kink configuration entropy is estimated to be around $5.23k_B$.

1. INTRODUCTION

In the development of modern materials theory there is a growing awareness of the importance of a fundamental understanding of the role of special lattice defects. For example, most physical properties of crystalline materials are affected more or less by the presence of dislocations and point defects and the interactions between them. In many cases these are dominated by the elastic strain field which, for distances more than a few atomic spacing from the center of the dislocation, is described well by linear continuum elasticity. However, it has been demonstrated[1] that the continuum description, which excludes the short-range details of important non-linear effects, cannot account for the intrinsic dislocation-lattice coupling which is the source of the anisotropic crystal plasticity in bcc metals. Therefore, a detailed description of the dislocation properties at atomic scale with proper boundary conditions has become increasingly important to advance the modern dislocation theory in crystal plasticity. The other important problem in atomistic-simulation studies of dislocations and other lattice defects is how to calculate the interaction potentials describing interatomic forces with sufficient accuracy. Specifically, it has been shown that the multi-ion *d*-state

interactions play an important role for bcc transition metals with partially filled *d*-bands, and therefore any proposed interaction scheme must explicitly contain angular-force contributions[2]. In this paper, we present our simulation results of fundamental atomic-level properties of point and line defects in bcc Ta using quantum-based multi-ion interatomic potentials derived from the model generalized pseudopotential theory (MGPT). Some validation tests of the MGPT potentials are presented in comparing point defect formation and migration energies with the *ab-initio* electronic-structure results and the experimental data. Robust 2D and 3D GF techniques have been used to simulate static and dynamic dislocation properties. In particular, the a/2<111> dislocation core properties under the influence of applied shear loading and temperatures were extensively studied using the MGPT-GF technique.

2. COMPUTATIONAL METHODS

2.1. *Ab-initio* electronic structures methods

For the point defect calculations, we used both full-potential linear muffin-tin orbital[3] (FP-LMTO) and plane-wave pseudopotential[4] (PP) techniques. Both methods are fully implemented within first-principles density functional theory (DFT). Because the PP method can handle larger number of atoms and perform structural relaxation in a much faster fashion, it is used to obtain relaxed defect structures as input into the FP-LMTO calculations. Such a combination of using two *ab-initio* techniques offers a very efficient and accurate approach for calculating defect structures and energetics.

2.2. MGPT interatomic potentials

The present calculations in Ta have been carried out using quantum-based, multi-ion interatomic potentials derived from model generalized pseudopotential theory (MGPT)[2]. This approach is based on the corresponding first-principles generalized pseudopotential theory (GPT)[5], which provides a rigorous real-space expansion of the total energy for a bulk transition metal in the form

$$E_{tot}(R_1...R_N) = NE_{vol}(\Omega) + \frac{1}{2}\sum_{i,j}{}' v_2(ij;\Omega) + \frac{1}{6}\sum_{i,j,k}{}' v_3(ijk;\Omega) + \frac{1}{24}\sum_{i,j,k,l}{}' v_4(ijkl;\Omega) \qquad (1)$$

, where $\boldsymbol{R}_1$ .... $\boldsymbol{R}_N$ denote the positions on the N ions in the metal, Ω is the atomic volume, and the prime on each sum over ion positions excludes all self-interaction terms where two indices are equal. The leading volume term in this expansion, E_{vol}, as well as the two-, three-, and four-ion interatomic potentials, v_2, v_3, and v_4, are volume

dependent, but structure independent quantities and thus transferable to all bulk ion configurations. The angular-force, multi-ion potentials v_3 and v_4 reflect contributions from partially-filled *d*-bands and are generally important for central transition metals. Within the MGPT framework[5], these potentials are systematically approximated by introducing canonical *d* bands and other simplifications to achieve short-ranged, analytic forms, which can then be applied to both static and dynamic simulations with the Green's function techniques described below. To compensate for the approximations introduced into the MGPT, a limited amount of parameterization is allowed in which the coefficients of the modeled-potential contributions are constrained by experimental or volume-dependent *ab-initio* theoretical data. The details of this parameterization for Ta, together with extensive validation tests of the potentials, are discussed in Ref. 1.

2.3. MGPT Green's function simulation method

For the study of dislocation properties, a method for dynamically updating the boundary conditions of atomistic simulations[6] has been used. The boundary conditions for 2D and 3D defect cells are evaluated using line[7] and point[8] force distributions, respectively. In this static and dynamic flexible boundary condition method, the simulation box is divided into three regions: atomistic, GF, and continuum. In the continuum region, the atomistic positions are initially determined according to the anisotropic elastic displacement field[7] for a dislocation line defect at the center of the atomistic region. Complete atomistic relaxation or updating is performed in the atomistic region according to the interatomic forces generated from Eq. (1). Forces develop in the GF region as relaxation or propagation is achieved in the atomistic region are then used to relax or propagate atoms in all three regions by the 2D or 3D lattice and elastic GF solutions for line[7] or point[8] forces.

3. DEFECT AND MECHANICAL PROPERTIES OF BCC TA

3.1. Vacancies and self-interstitials

Point defects, including vacancies, self-interstitials and impurities, can have an important impact on the mechanical properties of metals. In the calculations of vacancy and interstitial formation energies, we calculated the relaxed total energy $[E_{tot}(N, \Omega_N)]$ for a supercell with N atoms in a volume Ω_N and with periodic boundary conditions applied in all three directions. At zero pressure, the formation energy for the defect is defined

as $E^f = E_{tot}(N, \Omega_N) - NE(\Omega_0)$, where $E(\Omega_0)$ is the energy per atom of the bulk solid at the equilibrium atomic volume Ω_0.

Table I summarizes calculated vacancy and self-interstitial formation and migration energies and formation volumes for Ta using FP-LMTO, PP, and MGPT methods, together with available experimental data[9]. For the self-interstitial formation energy, only the <110> split-dumbbell configuration is presented here, which is found to be the lowest-energy configuration compared to other self-interstitial defects. The same conclusion is also found in bcc Mo[10]. The migration energy for the point defects is determined from $E^m = E^{saddle} - E^f$, where E^{saddle} is the formation energy of the defect at the saddle point. The optimized migration paths are found to be along <111> for both the vacancy and self-interstitials, with the latter corresponding to the so-called jump and rotation path. Overall, there is good agreement between theory and experiment and between the electronic-structure results and the MGPT results.

TABLE 1

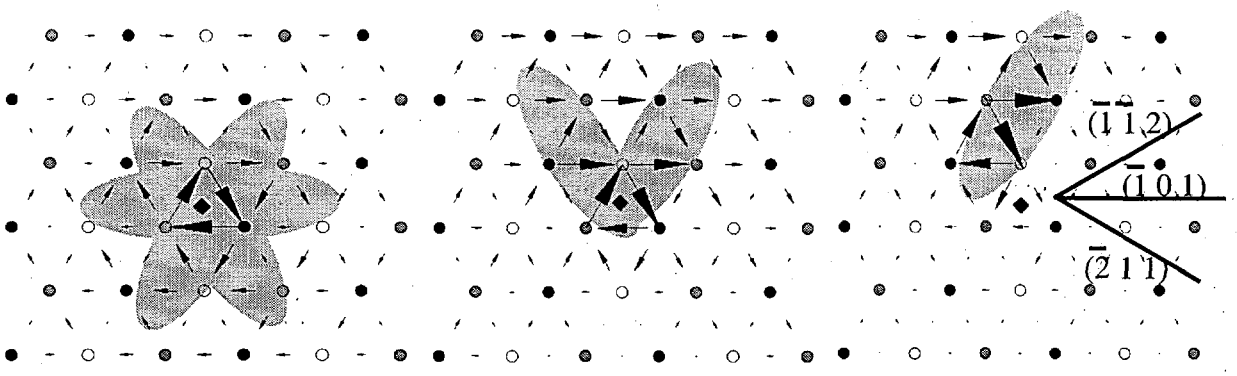
Vacancy and <110> split-dumbbell self-interstitial formation energies E^f_{vac} and E^f_{int}, migration energies E^m_{vac} and E^m_{int}, and formation volumes Ω^f_{vac} and Ω^f_{int} for bcc Ta. The MGPT vacancy and self-interstitial results were obtained from 250-atom and 1800-atom supercell calculations, respectively. The PP and FP LMTO vacancy results were obtained from fully relaxed 54-atom supercell calculations, using ten special k-points, except the FP LMTO vacancy migration energy where a 36-atom supercell was used. The PP self-interstitial results were obtained from corresponding 128-atom supercell calculations, which are slightly different from 48-atom supercell calculations reported in Ref. 1. Migration energies were calculated along <111> migration paths, as described in the text. The experimental data is from Ref. 9.

	E^f_{vac}(eV)	E^m_{vac}(eV)	Ω^f_{vac}/Ω_0	E^f_{int}(eV)	E^m_{int}(eV)	Ω^f_{int}/Ω_0
MGPT	2.89	0.78	0.53	6.30	0.50	0.40
PP	3.10	0.90	0.60	6.43	0.55	0.31
FP LMTO	3.10	0.74				
Experiment	2.8-3.1	0.7				

3.3. a/2<111> screw dislocation properties

The low-temperature and high-strain-rate plasticity of bcc metals is controlled by the intrinsic core properties of a/2<111> screw dislocations. Unlike the highly mobile edge dislocations in these metals, the motion of the screw dislocation is severely restricted by the nonplanar atomic structure of its core, resulting in low mobility and high Peierls stress. However, under the influence of external loading and temperature, the core

structure undergoes a transformation in the process of dislocation motion. In this section, we present our static and dynamic simulations of individual a/2<111> screw dislocations in bcc Ta using MGPT-GF technique.



(a) $\tau = 0$ (b) $\tau_{<111>(\overline{1}\overline{1}2)}=0.009G$ (c) $\tau_{<111>(\overline{1}\overline{1}2)}=0.012G$

FIGURE 1

The screw-component displacement maps of the dislocation core in Ta under applied pure shear stress in the <111> direction on the (-1 -1 2) plane. The <111> screw component of the relative displacement of neighboring atoms due to the dislocation is represented by an arrow between the two atoms. The symmetry of the core configuration is decreasing with increasing stress. . The applied stress is in units of shear modulus in the <111> direction, i.e. G = 0.625Mbar for Ta. The calculated Peierls stress is 0.0102G. The stress-free elastic center associated with the screw dislocation is indicated by a diamond symbol.

3.3.1. Stress-free ground-state core configuration

The stable ground-state core configuration of the a/2<111> screw dislocation is centered among three <111> atomic rows forming a triangular prism. Around these three rows the near-neighbor atoms are positioned on a helix that winds up in a clockwise or counter-clockwise manner, depending on the location of the elastic center and the sign of the Burgers vector **b**. In the present work, the atomic core structure in bcc Ta has been simulated using a 2D MGPT-GF technique in cylindrical geometry, with periodic boundary conditions along the <111> direction. Our results reveal that a nearly isotropic but still three-fold symmetric and doubly degenerate core, whose detailed structure is easily displayed using the differential displacement method[11]. The calculated screw-component map for Ta is shown in figure 1a. The MGPT dislocation core structure is comparable with the results obtained by other authors using different

potentials and using electronic-structure techniques[12], where their reported dislocation core structures show completely isotropic core configuration.

3.3.2. Core configurations under applied stress

When the straight dislocation is under external loading, the applied stress exerts the Peach-Koehler force on the a/2<111> screw dislocation. If the applied stress is large enough to overcome the Peierls barrier, the straight dislocation starts to move on the maximum resolved shear stress plane. During the course of loading with increasing stress, the dislocation core undergoes a transformation due to symmetry breaking induced by the external forces. In the present work, we only consider external loading corresponding to the application of a pure shear stress (τ) parallel to the Burgers vector and resolved on the (-1 -1 2) plane.

The effects of the applied stress on the dislocation core are shown in figures 1b and 1c. For stress levels somewhat lower than the corresponding critical resolved shear stress (τ_P), the core extends and spreads into (-1 1 0) and (0 -1 1) planes, resulting in a reduction of symmetry from nearly six-fold to two-fold. As the applied stress is increased slightly higher than τ_P, the dislocation starts to move by extending its core along the (0 -1 1) plane and constricting the other part of the core that is spread along the (-1 1 0) plane. This type of core transformation eventually reduces the corresponding core symmetry from two-fold to one-fold. The dislocation actually moves in a zigzag pattern on successive {110} planes, so the effective slip plane is in fact a {112} plane for bcc Ta.

3.3.3. Multiple-kink formation at finite temperatures

At finite temperatures, the straight dislocation is under the influence of thermal fluctuations and thus the possibility of forming double kinks is increased[13]. In the present work, we have considered finite-temperature GF simulation of dislocation motion under pure shear loading at a level about 10% below the calculated Peierls stress ($\tau_P = 0.0102G$). The simulation cell is constructed in a cylindrical geometry with periodic boundary conditions along the dislocation line direction. The atomistic region contains a radius of 40b (b=2.86Å is the length of the Burgers vector) and 200b in length. The simulation was performed at 300K.

Our simulation results reveal multiple-kink formation along the screw dislocation line. A snapshot of the atomistic kink structure is displayed in figure 2b. It is also possible to estimate the configuration entropy of forming thermal kinks by counting all possible double-kink configurations under various loading conditions. Statistically averaging over

35 loading conditions gives an average kink width of 3.7b and an estimate of the configuration entropy of about $5.23k_B$.

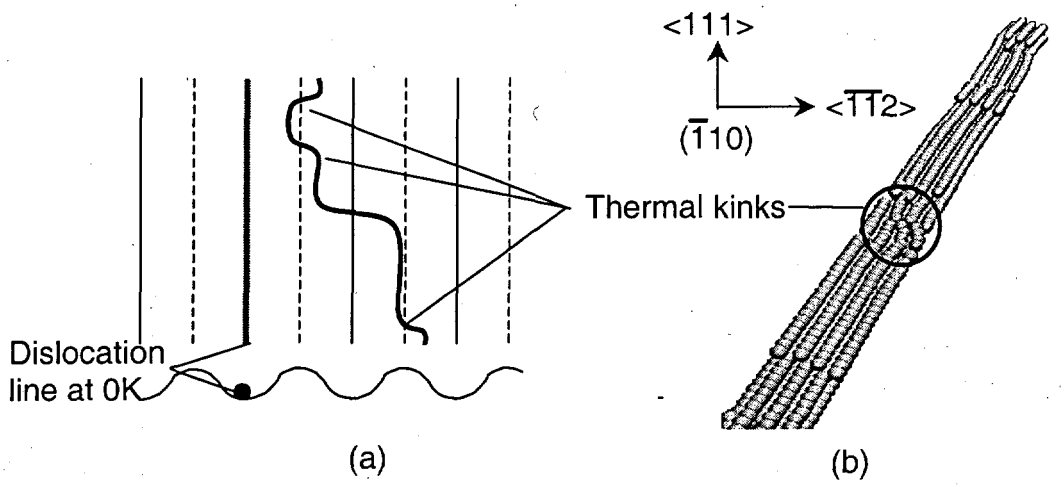


FIGURE 2

(a) Schematic drawing of thermal-kink formation at finite temperatures. (b) A snapshot of thermal-kink formation along the <-1 -1 2> direction on the (-1 1 0) plane.

4. DISCUSSION

In this paper, we have presented our work on quantum-based atomistic simulation of materials properties in bcc Ta. Central to this work is the development, from fundamental quantum mechanics, of robust many-body interatomic potentials for bcc transition metals via model generalized pseudopotential theory (MGPT), providing close linkage between *ab-initio* electronic structure calculations and large-scale static and dynamic atomistic simulations. Successful application and validation of MGPT potentials were demonstrated by comparing point defect formation and migration energies with the *ab-initio* electronic-structure results and experimental data. Robust 2D and 3D GF techniques have been used to simulate static and dynamic screw dislocation properties. Under external loading, our static simulation reveals a symmetry reduction in the dislocation core structure during the process of dislocation motion. At finite temperatures, the thermal fluctuations induce multiple-kink formation along {110} planes in our dynamic GF simulations. The configuration entropy associated with the thermal-kink formation is estimated to be around $5.23k_B$.

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